

Serial No. 09/831,915

DOCKET NO: 208608US0PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF

THOMAS DANIEL, ET AL.

SERIAL NO: 09/831,915

FILED: MAY 25, 2001

FOR: HYDROGELS CAPABLE OF
ABSORBING AQUEOUS FLUIDS

EXAMINER: METZMAIER, D. S.

GROUP ART UNIT 1712

DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes Dr. Manfred Essig who deposes and states:

1. That I am a graduate of the University of Kaiserslautern and received a Ph.D. degree in the year 1981.
2. That I have been employed by BASF SE (formerly BASF Aktiengesellschaft) for 23 years as a scientist in the field of Electron Microscopy.
3. That the following experiments were carried out by me or under my direct supervision and control.

OVERVIEW

The experiments relate to comparative compositions according to WO 97/46195 (Procter and Gamble).

Comparative Samples 1 (EDG-0007-17-A) and 2 (EDG-0007-17-B) as prepared by Dr. Braig (see the Declaration by Dr. Volker Braig) were investigated by SEM (Scanning Electron Microscopy) and EDX (Energy-dispersive X-ray spectroscopy) to show that the silicon corresponding to the alkali metal silicate is distributed on the surface of the superabsorbent particles according to WO 97/46195 (Procter and Gamble) and NOT inside the particles.

The results of the comparative examples show that there was NO silicon inside the SAP particles

SAMPLE DESIGNATIONS

Sample [1] was also designated as EDG-0007-17-A.

Sample [2] was also designated as EDG-0007-17-B

SAMPLE PREPARATION

In preparation of the measurement, gelatine capsules were filled with polymer particles of Samples 1 (EDG-0007-17-A) or 2 (EDG-0007-17-B), respectively, as prepared by Dr. Braig. The gelatine capsules were cylindrical with a diameter of about 8 mm. After the polymer particles of Samples 1 (EDG-0007-17-A) or 2 (EDG-0007-17-B), respectively, as prepared by Dr. Braig were already in the capsules, a polymer (commercially available LR-White, Agar Scientific Limited, Essex, England) was filled into the capsules. After hardening of the LR-White polymer in the capsules, two of the gelatine capsules were included in a resin cylinder with about 25 mm diameter. The cylinder was then cut with a diamond rotary cutter at about 20,000 rpm. The reason that two capsules were used for each sample was to increase the probability of cutting polymer particles more or less through the center when cutting the resin

cylinder. The cutting exposed a cross sections of the polymer particles of Samples 1 (EDG-0007-17-A) or 2 (EDG-0007-17-B), respectively. In order to make the surface electrically conductive, a thin film of carbon was deposited by vapor deposition on the cross section surface.

EXPERIMENTS

The experiments were performed using SEM and EDX. The SEM allowed generation of images of the cross-section surface of the sample and illustrated primarily the structure of the surface when employing secondary electrons. The exploitation of backscatter electrons gave information about the structure and in addition information about the density of a material at the sample surface.

The method of differentiating between different elements was EDX (Energy-dispersive X-ray spectroscopy). In the EDX method, atoms are excited with X-ray radiation. The excited atoms then emit X-rays which are specific for the respective element and which are measured.

A standard SEM method was used for the experiments. The cross-section sample as prepared was put on the stage of a SEM microscope (company LEO, model 1450VP). The standard imaging parameters were 5kV for the secondary electron images and 20 kV for the back scatter electron images.

For analytical purposes an EDX spectrometer was adapted to the scanning microscope. The system was from the company Noran, model SEX(NSS) *SEX FILE 04/01/2010*

A 20kV primary beam energy was used for the generation of the elemental distribution images obtained by EDX. In order to enhance the contrast when depicting the images with EDX in the report, only signal strengths are presented which were representative for local

Serial No. 09/831.915

concentrations above 50% of the average concentration in the sample. This focus on certain signal strengths improved correlation of the various elemental pictures and made the images easier to interpret.

IMAGES:

Two sheets with five (5) pictures each are attached to this Declaration and are incorporated into the Declaration by reference. One sheet is for comparative Sample 1 and another sheet is for comparative Sample 2. The analysis of the images was based on the most clearly visible particles of SAP. The field of view was representative for the whole cross section of the sample which was checked visually.

First sheet with five (5) pictures for Sample 1:

The first picture from the top shows an SEM image of the cross section of the sample without reference to any particular element.

The first picture from the top is a backscatter electron image and shows the location of higher densities brighter than the vicinity having lower density of the material of the sample cross section. The darkest areas (black) stand for the polymer used during preparation (commercially available LR-White, Agar Scientific Limited, Essex England) of the samples

The first picture from the top shows the spatial distribution of three granular components (SAP, zeolithe and sodium silicate) in the cross-section of the sample

The identification of the components goes along with the information in the elemental distribution images which are the remaining colored images on the first sheet.

- The large areas in dark grey from the first picture correspond to the bright yellow areas in the Na-image representing the superabsorber. The second picture from the top on the left side shows the distribution of sodium (Na)

- The bright areas in the first picture correspond to the medium-bright yellow areas in the Na-image as well as with the bright areas in the Si-image representing the alkali metal silicate.

- The distribution of medium-grey and dark-gray fine granulates correlates with the Al-image, as well as, partially, with the Si-distribution representing zeolitic particles.

- The black areas represent the LR-White.

The O-distribution is not relevant.

In summary there is no indication of Si distributed across the superabsorber particles.

Second sheet with five (5) pictures for Sample 2:

The first picture from the top shows an SEM image of the cross section of the sample without reference to any particular element. In other words, the first picture from the top is a backscatter electron image and shows the location of higher densities brighter than the vicinity having lower density of superabsorber. The darkest areas (black) stand for the polymer used during preparation (commercially available LR-White, Agar Scientific Limited, Essex England) of the samples

The first picture from the top shows the spatial distribution of two granular components (SAP, zeolithe) in the cross-section of the sample

Their identification goes along with the information in the elemental distribution images below:

- The large areas in dark grey of the first picture correspond to the bright yellow areas in the Na-image representing the superabsorber.

- The bright areas of the first picture correspond to the dark yellow areas in the Na-image as well as with the bright areas in the Si-image representing the alkali metal silicate. The second picture from the top on the left side shows the distribution of sodium (Na).

In addition there is also correspondence to the bright areas in the Al-image elucidating a sticking of the zeolite component to the alkali metal silicate.

The O-distribution is not relevant.

Silicon (Si) shown in the last picture at the bottom is not in the cross section of the polymer particle as shown by the black areas which correspond to the areas that are bright yellow or dark gray in the first and second picture on the left. Thus, there is NO Si inside the polymer particles.

RESULTS

The element distribution images for Sample 1 show that the SAP particles (recognizable by the high Na content in the images) and the zeolite phase (evident predominantly in the Al distribution image, since the zeolite used contains Al) as well as the sodium water glass phase (recognizable in the Si distribution image and the low Na content) are not present in association with one another. In Sample 1 there is only a physical mixture between the SAP, the zeolite and the solid sodium silicate. Both the sodium water glass and the zeolite contain Si and O, and so overlapping occurs in places in the element distribution images. This is also the case of Na in SAP and sodium water glass. In other words, there is NO Si to be observed within the particles

Serial No 09/831,915

The element distribution images of Sample 2 prove that the soda water glass phase and the zeolite phase have aggregated and are present either as separate particles or as crusts on the surface of the SAP particles.

The images prove that sodium silicate adheres on the SAP particle surfaces only, but is NOT distributed inside the polymer particles.

4. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing therefrom.

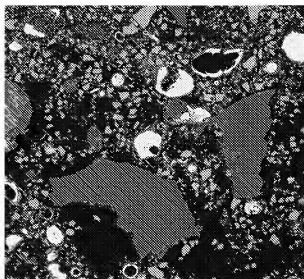
5. Further deponent saith not



Signature



Date



100 : 1

200µm

Auftrag: 114805

Abb: 01b100 RE

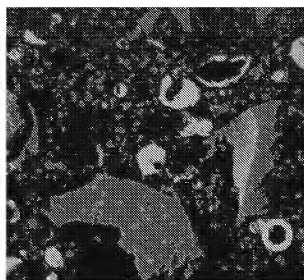


100 : 1

200µm

Auftrag: 114805

Abb: Na



100 : 1

200µm

Auftrag: 114805

Abb: O

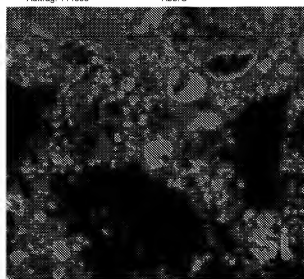


100 : 1

200µm

Auftrag: 114805

Abb: Al

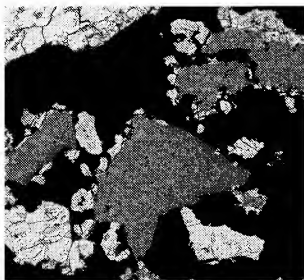


100 : 1

200µm

Auftrag: 114805

Abb: Si



100 : 1

200µm

Auftrag: 114805

Abb: 02b100 RE

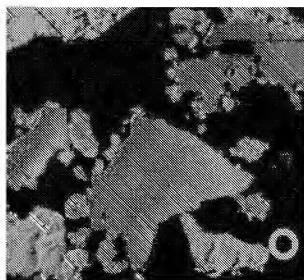


100 : 1

200µm

Auftrag: 114805

Abb: Na

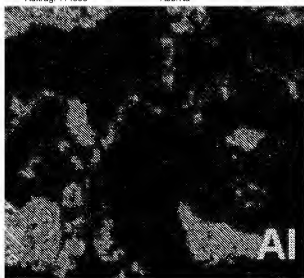


100 : 1

200µm

Auftrag: 114805

Abb: O



100 : 1

200µm

Auftrag: 114805

Abb: Al



100 : 1

200µm

Auftrag: 114805

Abb: Si